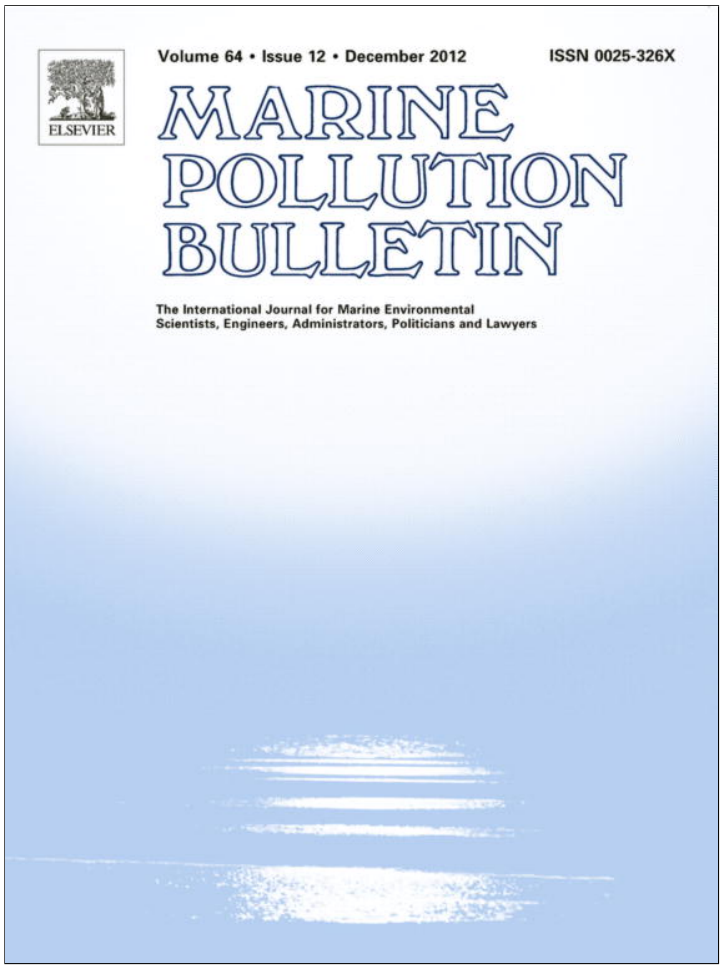

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Long-term decreases in persistent organic pollutants in South African coastal waters detected from beached polyethylene pellets

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ABSTRACT

Polyethylene pellets provide a convenient means to monitor Persistent Organic Pollutants (POPs) in marine systems. Pellets collected between 1984 and 2008 at three South African beaches were analysed for PCB, HCH and DDT. Concentrations of all three POPs decreased over the last two decades, although this signal was less clear for PCBs, and further monitoring is needed to assess trends in this family of compounds. DDT concentrations at two sites were higher than previous records for southern Africa, but there is no evidence of a link to the ongoing use of DDT for malaria control. HCHs concentrations were lower than in pellets from the east coast of southern Africa, suggesting that this pesticide was mainly used in the eastern part of the region. Our study demonstrates the potential for International Pellet Watch to track temporal as well as geographical patterns in the abundance of POPs in marine environments.

1. Introduction

There is growing concern about the ubiquitous nature of plastics in the environment (Thompson et al., 2009), especially in marine systems (Barnes et al., 2009; Ryan et al., 2009). One of their impacts is to transport a suite of persistent toxic pollutants and to introduce them into marine food webs when animals ingest plastic particles (Teuten et al., 2009). The tendency of hydrophobic organic pollutants to concentrate in plastic particles (Mato et al., 2001; Endo et al., 2005) can be used to monitor the distribution and abundance of bio-accumulative and persistent compounds in marine environments. The International Pellet Watch (IPW) was set up in 2005 to identify key source areas and potential violations of international regulations controlling the use of bio-accumulative and persistent compounds (Takada, 2006). It uses polyethylene pellets as convenient passive samplers to assess environmental loads of these compounds in coastal waters around the world. Polyethylene pellets are preferred because they are more efficient at accumulating persistent compounds than other plastic polymers such as polypropylene (Endo et al., 2005). The IPW recently published the first global map based on such samples, showing strong regional patterns in concentrations of different pollutants (Ogata et al., 2009; Teuten et al., 2009).

In addition to tracking geographical patterns in bio-accumulative and persistent compounds, repeated sampling of polyethylene pellets from the same site has the potential to track temporal changes in these compounds' concentrations in marine environments. The sorption/desorption of hydrophobic compounds to PE pellets is a slow process controlled by diffusion into the polymer matrix. Concentrations of compounds in pellets take 1–2 years to reach equilibrium with ambient concentrations in the sea (Karapanagioti et al., 2010). Thus, tracking historical changes in compound concentrations is limited to temporal scales of several years to decades. In this paper, we report decadal changes in PCB, HCH, and DDT concentrations for South African coastal waters based on historical collections of pellets dating back to the 1980s. These compounds are all listed in the Stockholm Convention on Persistent Organic Pollutants (POPs). We can therefore interpret the temporal changes of POPs concentrations in terms of international measures taken to stem the release of POPs into the environment.

2. Materials and methods

In 1984, PGR and CLM started a long-term monitoring programme to track changes in the abundance of litter stranded on 50 South African beaches (Ryan and Moloney, 1990). This included plastic meso-debris (2–20 mm fragments) collected by sieving a half-metre wide strip from the most recent high tide line up the beach to the storm strand line through a 2 mm-mesh sieve.

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Samples have been collected roughly every 5 years during the austral winter, and after enumeration, they are stored in glass vials or jars in a dark cupboard at room temperature. Following the call for pellets by the IPW (Takada, 2006), subsamples of pellets collected at three beaches that typically contain large numbers of pellets were submitted for analysis.

Pellet concentrations on South African beaches peak close to urban centres, suggesting that local sources are important contributors to adjacent beaches. One urban beach was sampled at Blue Water Bay (33°51′ S, 25°38′ E) in Port Elizabeth, the largest city in the Eastern Cape Province (Fig. 1). This beach lies just east of the Swartkops River estuary, the city's commercial port with a coal and petrochemical terminal. The Swartkops River passes through the main industrial area of Port Elizabeth, which supports a diversity of industries including plastics, carbon black, tires, and vehicle construction. As a result, many pellets collected here are likely to be of local origin. Pellet densities at Blue Water Bay have ranged from 1000 to 1800 pellets per linear metre of beach (Ryan and Moloney, unpubl. data).

Because we wanted to sample pellets that had been in coastal waters for some time, we also selected two rural sites where pellets tend to accumulate. One beach is on the west coast, on a remote stretch of 16 Mile Beach in the West Coast National Park (33°19′S, 18°08′E) near Yzerfontein, roughly 80 km north of Cape Town and 40 km south of Saldanha Bay (Fig. 1). Saldanha has a large steel factory commissioned in 2000. In surveys from 1984 to 2010, pellet densities at this beach ranged from 100 to 700 pellets per linear metre of beach (Ryan and Moloney, unpubl. data). The other rural site was at Woody Cape (33°46′S, 26°25′E), at the east end of Algoa Bay, a large, heart-shaped bay on South Africa's south coast. Woody Cape forms part of the Greater Addo National Park, but large concentrations of marine debris accumulate on the beach due an the eastward flowing inshore current which carries debris along the coast from Port Elizabeth and the newly constructed Coega harbour and industrial development that started construction in 2002. Pellet densities ranged from 1500 to 10,000 pellets per linear metre of beach from 1984 to 2005, mainly in the dune slacks behind the main beach area (Ryan and Moloney, unpubl. data).

Due to analytical constraints, samples collected at roughly decadal intervals were analysed from the three beaches: 1984, 1994 and 2005 for Woody Cape, and 1989, 1999 and 2008 for Yzerfontein and Port Elizabeth. We analysed three POPs: polychlorinated biphenyls (PCBs) and two organochlorine pesticides, hexachlorocyclohexanes (HCHs) and dichlorodiphenyl trichloroethane and its degradation products (DDTs). From each beach in each year, roughly 200 pellets were sorted into polyethylene, polypropylene and other plastic types using far-infrared spectroscopy (Plascan-W, OPT Research Inc., Tokyo, Japan). Among polyethylene pellets, those that appeared yellowed were selected for the analysis of POPs because they probably have been at sea for longer than whitish pellets and thus have had a greater opportunity to accumulate POPs (Ogata et al., 2009). For each year, five sets of yellowed pellets (each comprising five pellets) were analysed to estimate the representative concentrations for each of the three POPs and their congeners (see Ogata et al., 2009). However, the 1999 sample from the West Coast National Park contained too few yellowed pellets, so some non-yellowed pellets were included in these replicates.

Chemical analysis followed IPW protocols described in detail by Ogata et al. (2009), and consisted of extraction in hexane followed by column chromatography and analysis by gas chromatography linked to a mass spectrometer for PCBs and *p,p′*-DDE and to a electron capture detector (ECD) for *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDD, *o,p′*-DDD and HCHs. We measured concentrations of 13 CB congeners of PCBs ranging from tetra-CBs to nona-CBs, and for HCHs, we measured the concentrations of four isomers (α, β, γ, δ). Median concentrations of the five replicates are reported to reduce among-sample variance caused by differences in residence times in coastal waters between pellets or possible differences in the sources of individual pellets (Endo et al., 2005; Ogata et al., 2009). This is standard practice for the International Pellet Watch protocol, because the median values provided the best correlation

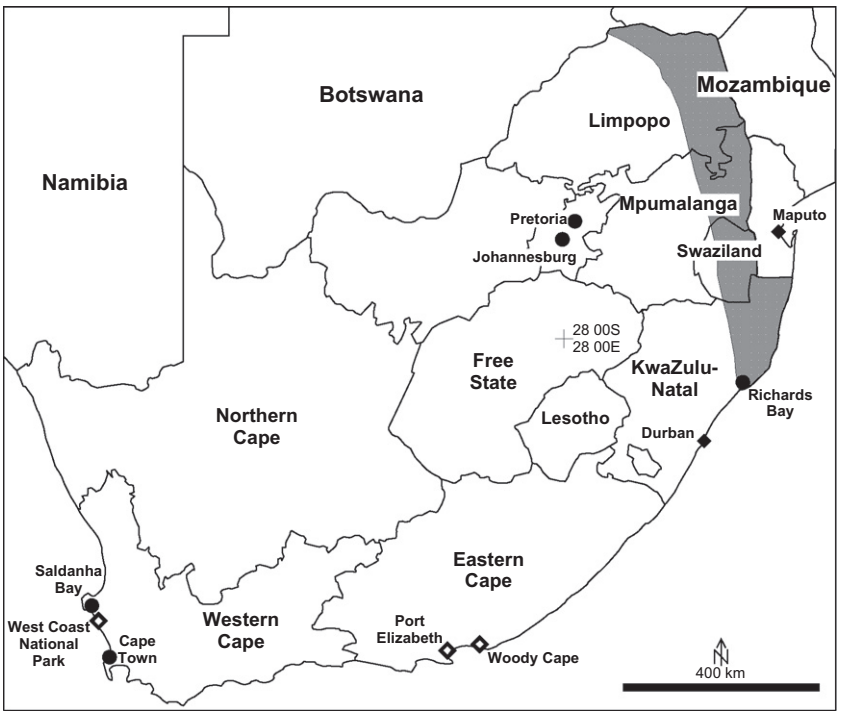

Fig. 1. The location of the three sampling sites in South Africa (open diamonds) in relation to other sites sampled by the International Pellet Watch (solid diamonds; see Ogata et al., 2009). Shaded area is where DDT is sprayed for malaria control.

with pollutant levels recorded in the same waters by Mussel Watch (Ogata et al., 2009).

3. Results

Of the pellets from each beach in each year, 82% were polyethylene, 11% polypropylene and 7% other polymers. The proportion of polyethylene pellets decreased over the sample period at all three beaches, averaging 86% in the 1980s and 1990s, decreasing to 77% after 2000 ($\chi^2 = 27.7$, d.f. = 2, $P < 0.001$). Slightly less than half of the polyethylene pellets were classified as 'yellowed'.

Overall, median concentrations of all POPs in pellets decreased from the 1980s to 2000s (Table 1 and Fig. 1), although variances among replicates were large in some samples (e.g., DDTs for West Coast Park in 2009; Fig. S1). Only pellets from Port Elizabeth showed consistent decreasing trends over all three periods, while the POP concentrations in pellets from rural sites increased for ΣPCB and ΣDDT from the 1990s to 2000s. ΣHCH concentrations showed the clearest decreasing trend, with consistent decreases throughout the sampling period at all three beaches. Although ΣHCH concentrations in the 1980s varied considerably among beaches, concentrations post 2000 were uniformly low (2–5 ng/g-pellet, Table 1). The γ isomer dominated the HCH composition in all samples (Table 1).

Absolute concentrations of ΣPCB were greater on the west coast than on the south coast in both in the 1980s and 2000s, suggesting that Cape Town and/or Saldanha Bay may have been a more significant point source than Port Elizabeth. Among the congeners of PCBs, the lower-chlorinated congeners were proportionally less in 1999 and 2008 (Table 1).

Concentrations of ΣDDT also were appreciably greater on the west coast than on the south coast in the 1980s, but have decreased substantially over the following two decades. By comparison, decreases have been less marked at the two south coast beaches, with a slight increase in ΣDDT in pellets from Woody Cape from 1994 to 2005 (Table 1). However, the concentrations of ΣDDT from 1999 to 2008 at all beaches ranged between 8 and 31 ng/g pellet. In contrast with the general decline in ΣDDT, the proportion of *p,p′*-DDT and *o,p′*-DDT relative to ΣDDT (%DDT in Table 1) ranged in a relatively narrow band between 59% and 73% with no discerning temporal trend at any of the beaches.

4. Discussion

Widespread use of PCBs and DDT was banned in most countries during the 1970s, and HCH from the 1980s to 2000s, but there is scant information on temporal changes in the use and environmental levels of most of these compounds in South Africa's marine environment against which we can judge our findings. Taking into account that different investigations quantify different congeners, ΣPCB concentrations in our recent samples (25–61 ng/g-pellets) are similar to the value reported from pellets collected near Durban (41 ng/g-pellets), the other major industrial centre on the South African coast, and are higher than the global background pollution level (*ca* 10 ng/g; Heskett et al., 2012) which are typical of many developing country samples (Ogata et al., 2009). The roughly 50% decrease found over the last two decades suggests that the ban on PCB usage has been moderately effective in reducing environmental burdens of these compounds. The faster decrease in lower-chlorinated PCB congeners could result from the preferential degradation and/or evaporation of lower-chlorinated biphenyls (Erickson, 1997). The apparent increases in PCB concentrations from the 1990s to 2000s at the two rural beaches may be cause for concern. For West Coast Park, this increase might be due to an underestimate of PCB concentrations in the 1999 sample because not all pellets used were 'yellowed'. However, the recent start-up of industrial activities linked to major port developments at Saldanha Bay (west coast) and Coega (near Port Elizabeth) also may explain the local increases in PCBs from a combination of

Table 1
Temporal changes in the concentrations of three compounds (ng/g-pellet) and their congeners sampled from yellowed polyethylene pellets collected at three South African beaches.

Compound	West Coast Park			Port Elizabeth			Woody Cape		
	1989	1999	2008	1989	1999	2008	1984	1994	2005
CB-66	15	1	0	2	1	<1	1	<1	<1
CB-101	8	1	1	4	2	1	4	1	1
CB-110	11	2	2	6	3	3	6	2	2
CB-118	8	<1	1	1	1	<1	2	<1	<1
CB-105	4	<1	1	<1	<1	<1	1	<1	<1
CB-149	11	2	5	5	5	4	8	2	4
CB-153	17	2	9	5	5	5	10	2	3
CB-138	23	4	19	11	14	8	16	6	9
CB-128	2	<1	2	2	1	1	2	1	1
CB-187	3	1	6	1	1	1	2	<1	1
CB-180	6	1	9	1	1	1	2	1	1
CB-170	5	1	6	1	2	1	2	1	1
CB-206	n.d.	n.d.	<1	n.d.	n.d.	n.d.	<1	n.d.	n.d.
ΣPCB	113	16	61	41	36	27	56	16	25
α-HCH	2	<1	<1	2	<1	<1	1	<1	<1
β-HCH	1	n.d.	n.d.	1	1	1	<1	<1	<1
γ-HCH	107	30	3	17	10	4	4	3	2
δ-HCH	1	n.d.	n.d.	<1	<1	<1	<1	<1	<1
ΣHCH	112	31	3	20	11	5	5	4	2
p,p′-DDT	695	7.9	19	10	6.3	2.4	33	12	19
o,p′-DDT	67	3.2	2.4	1	1	1.6	4	3.3	1.1
p,p′-DDE	241	1.4	1.9	1.4	0.6	0.3	0.8	1.9	3.9
p,p′-DDD	47	3.1	2.6	1.1	0.7	1.8	5	2.3	2.3
o,p′-DDD	231	2.4	4.7	4.1	2.1	1.9	14	1.6	3.6
ΣDDT	1281	18	31	18	11	8.0	57	21	30
%DDT*	59	62	70	63	68	50	65	73	67

n.d. = not detected. Concentrations are median values from a series of five pooled samples

* Percentage of ΣDDT comprised of *p,p′*-DDT and *o,p′*-DDT.

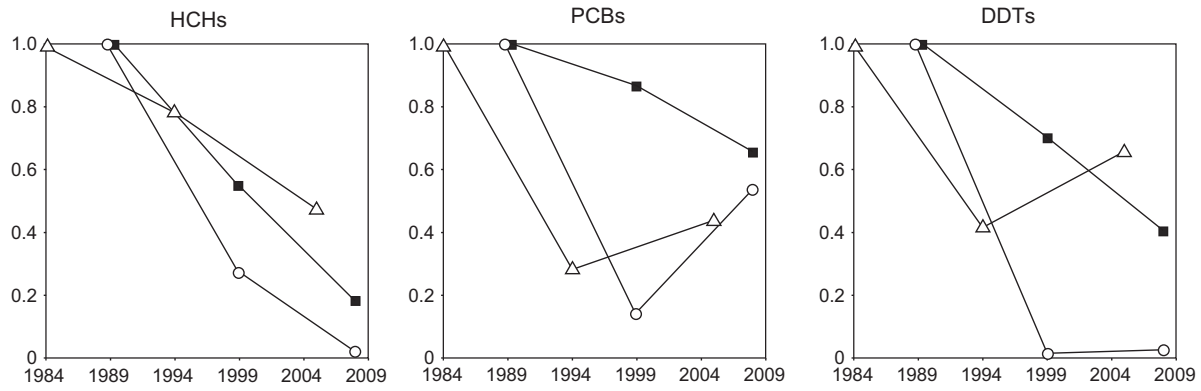

Fig. 2. Temporal changes in the relative concentrations of the three families of POPs on virgin pellets collected at three South African beaches: West Coast National Park (open circles), Port Elizabeth (solid squares) and Woody Cape (open triangles). Data are standardised relative to the maximum concentration at each site.

re-suspension of sediment during harbour construction and maintenance dredging, as well as steel manufacturing starting up at Saldanha. The Saldanha steel plant uses gas from coal gasification rather than cokes, but probably still results in the formation of PCBs from sintering and other high temperature processes (Beukens et al., 2001). SPMD (semi-permeable membrane devices using polyethylene) devices left in the water for 10 weeks at Saldanha in 2009 (Degger et al., 2011, and additional data) contained 30 μg/g ΣPCB (CB-29 = 3.6; CB-77 = 2; CB-138 = 24 μg/g; other PCBs were below quantification limit). The SPMD PCB profile was weighted towards the heavier PCBs as it was for the 2008 pellet profile from the adjacent West Coast National Park (Table 1). SPMDs exposed at Port Elizabeth in 2009 (Degger et al., 2011 and additional data) contained 105 μg/g (CB-18 = 8; CB-28 = 19; CB-29 = 19; CB-44 = 7; CB-52 = 4; CB-66 = 2.5; CB-77 = 11; CB-138 = 43 μg/g; other PCBs were below detection limit), which, as far as can be judged, seems similar to the pellet profile (Table 1). It would be prudent to undertake further monitoring of trends in this family of compounds as legacy sources (such as use of PCBs in transformers) may now be supplanted by current industrial sources.

Like PCBs, DDT persists in the environment, only slowly disappearing through degradation, evaporation, and burial in sediments. DDT was banned for general use in South Africa in 1974, but limited use of DDT to combat malaria continued in the northeastern part of South Africa until 1995, and resumed in 2000 (Wells and Leonard, 2006) following an increase in the incidence of malaria in the northwestern parts of the country (Fig. 1). Currently, some 20–40 tons per year is used in anti-malarial operations in South Africa and Swaziland (Wells and Leonard, 2006, G. Verdoorn, Griffon Poison Information Centre, pers. comm.). If these malaria control operations were a significant source of DDTs in regional coastal waters, we would expect levels to peak farther up the east coast (Fig. 1), as runoff from the area where DDT continues to be used enters the sea in Mozambique and northern KwaZulu-Natal. This is not the case; concentrations on pellets at both Yzerfontein and Woody Cape in the 2000s were appreciably greater (25 ng/g-pellets) than those from pellets sampled in Durban and Maputo in 2007 (Ogata et al., 2009), or at Port Elizabeth in 2008 (all 2–4 ng/g-pellets). Stockpiles of DDT continued to be used illegally in agricultural applications into the 1990s (Wells and Leonard, 2006), possibly accounting for the elevated levels detected in pellets. However, DDT concentrations in apex consumers such as vultures decreased dramatically from the late 1970s (Mundy et al., 1982) to late 1990s (van Wyk et al., 2001), suggesting that the agricultural use of DDT fell substantially over this period.

The high %DDT found in the pellets (59–73%; Table 1) also is difficult to explain. Similar high values were recorded in fish collected at Port Elizabeth between 1976 and 1979 (69%; Gardner et al., 1983) shortly after the ban on the agricultural use of DDT in South Africa, but subsequent samples have all had much lower %DDT values (e.g. tissue samples from seabirds and sharks all had values <3 %DDT; Schlenk et al., 2005; HB unpubl. data). Only sediments from South African harbours collected in 2007 (14–35 %DDT; Roos et al., 2011) have approached the pellet values. It thus appears that the high %DDT found in our pellet samples do not reflect the proportions of DDT isomers in current biological samples, but more closely reflect sediment samples. The DDT used for malaria control is made up of about 72% *p,p′*-DDT and 22% *o,p′*-DDT (Bouwman et al., 2006) and is thus unlikely that its past or recent use explains the high%DDT in pellets unless there are as yet unexplained isomer-specific kinetics or dynamics of the different DDT isomers and breakdown products.

Compared to PCBs and DDTs, HCHs are much less persistent in the environment (Ogata et al., 2009). The rapid decreases in concentrations of HCHs in pellets from South Africa is best explained by decreasing usage of this pesticide over the last two decades. HCHs were found in substantial concentrations at only a few sites in the world during the initial IPW findings (Ogata et al., 2009), with by far the highest loads recorded from the two sites sampled on the southern African east coast. The values recorded from our most recent samples were an order of magnitude lower (2–5 ng/g-pellets) than Durban and Maputo (34–37 ng/g-pellets; Ogata et al., 2009), but they remain elevated relative to most other regions of the world, indicating unknown current sources and scope for further improvement. Ogata et al. (2009) identified Lindane, a pesticide that contains mainly γ HCH, as the most likely source of this material. The γ isomer of HCH also dominated samples in our study, strongly indicating its use as an insecticide and not HCH production, which has a much higher proportion of α- and β-HCH. Lindane was only banned in SA in 2009 due to its listing in the Stockholm Convention. All registrations were voluntarily withdrawn by industry. However, illegal use of Lindane (probably from left-over stock) continued in some South African agricultural sectors after the ban (G. Verdoorn, Griffon Poison Information Centre, pers. comm.), and the ban against its use needs to be strictly enforced. It is also possible that countries north of South Africa may still be using left-over stock. However, the steady decrease in ΣHCH seen at all three beaches (Fig. 2) may be more indicative of its decreasing global and local use that started before 1994 due to other pesticide classes such as pyrethroids being introduced.

In general, concentrations of the three POPs analysed from pellets have decreased at all three sites sampled, suggesting that the amounts in South African coastal waters have decreased over the last two decades. Our results support the value of routine sampling of polyethylene pellets to monitor changes in environmental burdens of POPs in coastal waters, as proposed by the International

Pellet Watch (Takada, 2006). However, very little is known about the compounds that have been added to the Stockholm Convention since 2004, and monitoring their temporal trends in pellets from the southern oceans is recommended.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2012.09.013.

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